

Study of Pu Ion Productions in Thermal Ionization Mass Spectrometry for Plutonium Isotopic Ratio Measurements

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Accurate and precise analyses of ultra low-level Plutonium (Pu) and its isotopic composition are critically needed in study of Pu transport in environment, in-vitro radiobioassay, nuclear site remediation, and implementation of both nuclear test-ban and non-proliferation treaties. Mass spectrometry has significant advantages over alpha spectrometry for determining the quantity and the origin of Pu isotopes. By actively producing Pu ions through thermal ionization rather than passively detecting Pu alpha decay, mass spectrometry is capable of analyzing $\sim 10^6$ atoms of Pu within an hour after separated from sample matrices, thereby allowing for a faster and more sensitive measurement of Pu. Additionally, alpha spectrometry doesn't differentiate between the 239 and 240 Pu isotopes nor is able to measure the 241 Pu isotope since it is a beta emitter. However, since Pu isotopes are resolved according to their mass-to-charge ratios in mass spectrometer, quantitative analysis of ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu, and their ratios can be obtained from a single mass spectrometric measurement.

Measurement sensitivity, accuracy, and precision depend critically on the production of Pu ions from the surface of the rhenium (Re) filament in Thermal Ionization Mass Spectrometry (TIMS). Previous studies have shown that the method of depositing Pu on to Re metal surface directly affects the efficiency, stability, isotopic fractionation, and duration of Pu ion production; therefore, the sample deposition method needs to be optimized at low-level Pu measurement. The three deposition methods that will be evaluated are direct deposition, electroplating, and resin bead loading. The characteristics affecting the Pu ion production will be compared at the picogram level for each method with a certified Pu isotopic standard of CRM-137. Additionally, the resulting best method will be used to measure the massic activity and isotopic composition of Pu in the NIST bone ash standard reference material (SRM-4356), for which the results will be presented.